Third United Nations International Conference on the Peaceful Uses of Atomic Energy

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Abstract

PHASE EQUILIBRIA BETWEEN INTERMETALLIC COMPOUNDS IN URANIUM ALLOYS

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Intermetallic uranium compounds are of great practical importance:

1. As constituents in most uranium alloys.
2. As fissionable substances dispersed in metals of suitable nuclear properties.
3. As reaction products between uranium-containing fuels and can materials.
4. As potential fuel materials.

In all cases the intermetallic compounds influence the properties of uranium alloys, cans, and matrix materials of dispersion-type fuel elements. Therefore, the practical use of uranium-containing alloys required a good knowledge of constitution, properties, phase relations, and stabilities of intermetallic uranium compounds. Careful constitutional investigations combined with corresponding measurements of the most interesting properties are normally the first step in establishing the desired knowledge. To this a contribution is given in the present paper.

More than thirty phase diagrams of multicomponent uranium alloy systems have been studied by us. Thereby relatively often quasibinary, quasiternary etc. phase relations, and some ternary uranium compounds have been found.

Under consideration of all known data in this field, the quasibinary systems can be divided into three types:

Type I: Systems between intermetallic uranium compounds.

Type II: Systems between one intermetallic uranium compound and one of the pure components.

Type III: Systems between one intermetallic uranium compound and one intermetallic uranium-free compound.

All these quasibinaries are simple eutectic systems if the boundary compounds exhibit different crystal structures (for example: U_3Si_2-UAl_2, Mo-UAl_2 $ZrAl_2$-UAl_2, UIr_2-URe_2 etc.). Generally, the two-phase regions in these eutectic systems extend over a wide range of concentration and temperature.

In quasibinary systems having boundary compounds with the same crystal structure complete solid solubility (for example: UFe_2-UIn_2, UFe_2-UOs_2, UCo_2-UIr_2 etc.) or ternary uranium compounds (for example: UFe_2-UNi_5, UAl_2-UNi_5, UAl_2-UFe_2 etc.) have been observed. So far 12 new ternary compounds have been found and determined.

It is possible to give a systematic arrangement of the experimental results in terms of valency electron concentration which in turn allows some predictions of the constitution of the quasibinaries and the nature of the ternary phases. Furthermore, on this basis the identification of unknown intermetallic uranium phases in technical alloys is possible.

Preparation, workability, casting properties, melting points, thermal expansion, corrosion and transformation behaviour, and other properties of some quasibinary uranium alloys and ternary compounds have been examined. It has been found that some of these alloys exhibit remarkable properties which make them attractive as potential fuel materials.

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Abstract

THE PRESENT STATE OF KNOWLEDGE ABOUT CADMIUM- AND INDIUMTANTALATE AS POSSIBLE CONTROL MATERIALS FOR HIGH-TEMPERATURE REACTORS

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Principally cadmiumtantalate and indiumtantalate can be used as absorber materials in high temperature reactors. Compared with boron or boron-containing materials which have almost exclusively been proposed till now for higher temperatures, the tantalates have the advantage in absorbing neutrons by a n,γ-reaction instead of a n,α-reaction. In relation to hafnium, the lanthanons or their oxides or other compounds, which are also interesting materials for higher temperatures, the tantalates are easily available at relatively low costs.

Mixtures made of cadmiumtantalate and indiumtantalate possess an advantageous absorption spectrum for neutrons as well in the thermal as in the epithermal energy region. This spectrum is comparable with that of hafnium or of the silver-indium-cadmium alloys. Furthermore the double-oxides are, with respect to tantalum, also interesting for fast reactors since only the isotope B^{10} can compete with tantalum in that energy range (0.01 MeV up to some MeV).

$Cd_2Ta_2O_7$ has good thermal resistance up to 1000°C, $In_2Ta_2O_8$ even up to 1300°C. The tantalates do not show any phase transformation between room temperature and 950°C (the highest temperature that was investigated). The double-oxides can be prepared easily by annealing the pure components CdO and Ta_2O_5 or In_2O_3 and Ta_2O_5. Cadmiumtantalate is compatible with silver and copper and, up to 700°C, with nickel; indiumtantalate is completely compatible with silver, copper and nickel and up to 700°C also with molybdenum and to some degree with iron. These results are in agreement with thermodynamical calculations.

For an estimation of the behaviour of the materials under reactor conditions the daughter products which originate by neutron absorption have to be considered. Whereas Cd^{113} transforms into the stable Cd^{114}, tantalum is transmuted into tungsten and indium into tin. Both daughter products can bind more oxygen in their most stable valency states than the parent elements. Therefore the reduction of Cd^{++} to metal can be expected while indiumtantalate should be stable. This has been confirmed by experiments with SnO- and WO_2- additions to cadmiumtantalate. Addition of copper (I)oxide to the compounds can suppress this effect.

For the practical use of the tantalates their workability of the pure compounds as well as of a cermet is of importance. While sintered pellets of the pure tantalates which were originally pressed at room temperature cannot be shaped by plastic deformation, there is no difficulty in deforming tantalate-copper cermets in a suitable can up to high degrees. Even after high degrees of deformation the distribution of the tantalate particles remains even, no clustering occurs. This even distribution is not disturbed by a recrystallization treatment of the deformed cermet.

Investigations of the behaviour of the tantalates under reactor conditions are initiated.

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PROPERTIES AND TECHNOLOGY OF ZIRCONIUM ALLOYS WITH NIOBIUM AND TIN

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Since some years zirconium alloys with niobium have been of interest in respect of their use as cladding and structural materials for water-cooled nuclear reactors. Three research contracts of the European Atomic Energy Commission (EURATOM) with Metallgesellschaft AG dealt with factors influencing the behaviour of zirconium alloys with niobium and tin.

The corrosion resistance of many zirconium alloys containing up to 5% niobium and up to 1.5% tin with additions of palladium, chromium and molybdenum was investigated by exposing the specimens to 350°C water and 400 and 480°C steam. It was found that the heat treatment has a great influence on the corrosion behaviour. With an appropriate heat treatment the zirconium niobium alloys have nearly the same corrosion resistance as zircaloy-2. The best corrosion resistance was attained by annealing below the α/β-transformation range which was investigated by constitutional studies at 580°C $\pm$ 10°C. With the appropriate heat treatment the corrosion resistance is nearly independent of the niobium content. Annealing above 600°C causes the appearance of β_{Zr} which has an unfavourable effect on the corrosion behaviour. Tin additions have a favourable effect at 350°C, no effect at 400°C and an unfavourable effect at 480°C.

The hydrogen pick-up of niobium containing zirconium alloys during corrosion is less than it is found in zircaloy-2.

Due to their hot strength alloys with more than 2% niobium are superior to zircaloy-2. Especially an alloy with 3% niobium and 1% tin showed excellent corrosion resistance and hot strength. The tin content is necessary to have a good strength at temperatures up to 450°C. This alloy was studied more intensively.

A hydrogen content in the alloy ZrNb3Sn1 favours a martensitic structure with an increase of hardness. A hydrogen content up to 1000 ppm has no bad influence on the creep strength. Impact tests showed that hydrogen-containing and hydrogen-free samples of ZrNb3Sn1 are superior to the corresponding samples of zircaloy-2.

It is possible to fabricate seamless tubes of ZrNb3Sn1. The fabrication possibilities are discussed. The alloy may be welded by normal welding procedures, used for zirconium, but it is necessary to heat treat the welds to improve their corrosion resistance.

The influence of a neutron radiation on the properties of ZrNb3Sn1 was tested in the Geesthacht reactor FRG 1 of the Gesellschaft für Kernenergieverwertung in Schiffbau und Schiffahrt mbH. Samples for testing the radiation effect on the tensile and impact properties and on the structure were kept in the coolant of the reactor. Corrosion tests under irradiation were performed in heatable autoclaves within the reactor. The maximum radiation dose was $8 \cdot 10^{19}$ nvt. The results of the radiation tests are discussed.

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Prospecting for and Dressing of Uranium Ore in the Federal Republic of Germany: Progress in Methodology and New Results

by

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The technical improvement of radiometers in Germany has currently followed the general trend of measuring techniques. Thus, for example, the digital recording and processing of data gathered in airborne measurements is well advanced. Geochemical uranium prospecting with mobile laboratories has been improved to such an extent that a small field crew can carry through 1,500 soil and water analyses per month. The modern methods of mineralogy including X-ray and structure analysis have rendered predictable from surface outcrops the mineral associations at greater depths. Absolute age determinations on ores and rocks by various methods, together with gamma spectroscopy have proved to be an important auxiliary means for judging the genesis of deposits. Particularly successful was the prospecting in the southern part of the Black Forest, where a pitchblende-bearing deposit was discovered in red iron-baryte-quartz veins. In large parts of the deposit concentrations of greater than 1 % U were found. Owing to the extremely low present world market price of yellow cake, extensive research and technical development aimed at a lowering of production costs in the dressing of high-grade and low-grade ore. Relevant investigations using ores coming from 10 different deposits have been carried out in the dressing plant of Ellweiler. Only recently final tests were performed on a method which by a combination of ion exchange and liquid-liquid extractions permits recovery of nuclear pure solutions and uranates in the course of uranium-ore dressing.

Examples for the topics outlined are presented.